Nuclear Data Scoping Studies for Nonproliferation

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ABSTRACT

Two nuclear data scoping studies were initiated to determine the impact of nuclear data to the nonproliferation mission and to suggest a path forward to resolving important nuclear data issues. The first study is a comprehensive review of (α,n) nuclear data. The (α,n) study examined the uncertainties in (α,n) reaction neutron yields and attempted to quantify the impact to nonproliferation applications. A primary focus was the $^{19}F(\alpha,n)^{22}Na$ reaction, but (α,n) data for actinides mixed with oxides, salts and other light isotopes were also examined. The nuclear data needs were prioritized, and a science plan was developed to resolve the most impactful deficits in nuclear data. This plan includes: energy integral and differential measurements on thin and/or thick targets; benchmarking of α particle stopping powers in varying matrices; neutron- and gamma-emission spectrum measurements; new (α,n) reaction evaluations; and modernization of codes used to predict the source term. The second scoping study examined the nuclear data required for active neutron interrogation. This study examined fast-neutron gamma-ray production data, evaluated the state of the nuclear data and the current modeling capabilities for neutron induced gamma-ray production, and it identified important signatures that may benefit nonproliferation applications. Several nonproliferation measurement systems rely heavily on the detection of gamma rays from neutron-induced reactions in interrogated objects, but neutron-induced gamma-ray production data are incomplete or inaccurate for many isotopes of interest. This study determined how the incomplete data limit the ability to model these systems. The nuclear data needs were prioritized, and a science plan was developed to resolve the most impactful deficits in nuclear data.

INTRODUCTION

The need for (α,n) nuclear data and gamma ray production from active interrogation applications was identified during recent nuclear data workshops, prompting two scoping studies to examine the uncertainties introduced by these data in nonproliferation applications. The goal of these studies is to provide actionable plans to resolve nuclear data issues by identifying needs, quantifying impact to applications and identifying goal uncertainties. This paper will provide an overview of the applications impacted, the nuclear data needs and recommendations.

(α,n) SCOPING STUDY

The (α,n) scoping study examined all aspects of nuclear data needs for applications that require information on the neutron and/or gamma emission from (α,n) reactions. The study examined the state of the nuclear data and the codes used to predict the (α,n) neutron sources. The neutron emissions from (α,n) reactions are important for many safeguards, nonproliferation and nuclear reactor applications. Due to the fact that UF_6 is the most abundant material in the fuel cycle, small uncertainties in the (α,n) neutron yield can result in several significant quantities of unaccountable material [1]. In another study of plutonium oxide, it was determined that the (α,n) source term was the primary source of uncertainty in the determination of Pu mass [2]. Advanced reactors require knowledge of the (α,n) source term on light materials in new fuel materials such as SiC, and FLiBe, MgCl and other molten salts.

The (α,n) data: The current ENDF (α,n) sublibrary contains one set of data for ^{4}He with new evaluations for ^{17,18}O and ^{9}Be from the Naval Nuclear Laboratory ready to be included in the next ENDF release [3]. Two other libraries exist: the JENDL-AN/2005 library [4], which is the Japanese special-purpose (α,n) reaction nuclear data library, and TENDL [5], which is a theory-based library. There are, for a number of (α,n) reactions, substantial differences between the different libraries and between the libraries and experimental data. For example, a recent oxygen evaluation (Fig. 1) revealed differences in the neutron emission spectra between data libraries sufficient to impact inferences using the measured total neutron yield. The large low-energy peak present in the JENDL/AN-2005 results can be attributed to the inappropriate use of Kalbach-Mann systematics in the ^{17,18}O evaluations [6]. The majority of the nuclides evaluated in JENDL-AN/2005 use Kalbach-Mann systematics, raising questions regarding its suitability for use in the targeted applications. Establishing a new ENDF sublibrary for (α,n) is an important step to support SOURCES4C [1] as well as transport codes such as GEANT4 [7], MCNP6.2 [8], MC21 [9], and COG [10]. Improved measurements and modern evaluations including the neutron and gamma yields and spectra are also critical for priority isotopes.

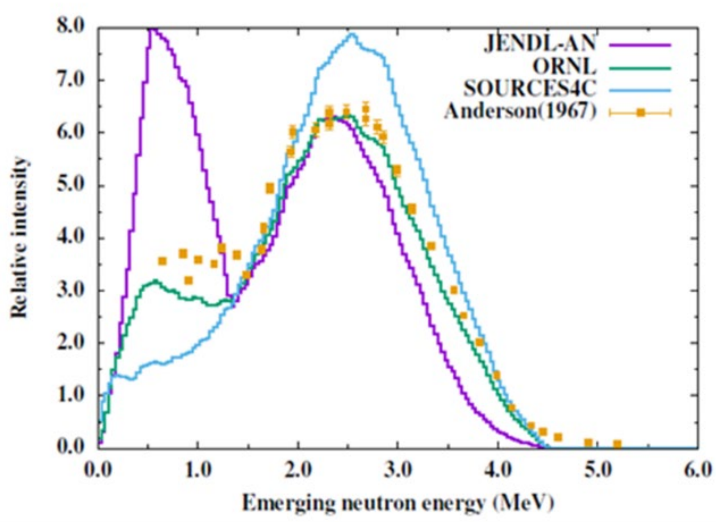

Figure 1 Theoretical calculations of the neutron spectrum from O(α,n) reaction compared to the JENDL-AN/2005 library, SOURCES4C, and experimental data [Pigni, 2016].

Modeling and simulations: SOURCES4C is one of the most widely used codes to calculate the neutron source from (α,n) reactions. It is distributed through The Radiation Safety Information Computational Center (RSICC) but is not maintained. SOURCES4C uses a hard-coded set of (α,n) cross section libraries that were incorporated by the developer. These data and the stopping powers have been edited for specific projects, but not included in an updated and distributed version of the code. Users would benefit from a modernization of SOURCES4C that interacts with codes such as MCNP, ORIGEN [11] and other transport codes through an application programming interface (API) and obtains its data from the latest version of a new ENDF (α,n) sublibrary.

A sensitivity study was undertaken to examine the impact of $^{19}F(\alpha,n)$ nuclear data uncertainties and evaluate the sensitivity of nondestructive verification measurements to the $^{19}F(\alpha,n)$ energy spectrum when using the Passive Neutron Enrichment Meter (PNEM) instrument. The $^{19}F(\alpha,n)$ data impact was compared to other measurement uncertainties such as the distribution of the UF_6 within the cylinder. An illustration of the measurement system is shown in Figure 2. The conclusion of the study was that the largest source of uncertainty in the accuracy of simulations was the total neutron yield from (α,n) reactions, where the reported thick target yields have a reported uncertainty of 7.3% [12] to 1.1% [13]. This impact was determined to be larger than the corresponding impact from uncertainty in the neutron spectrum due to the limited sensitivity of the PNEM to the lower energy neutrons.

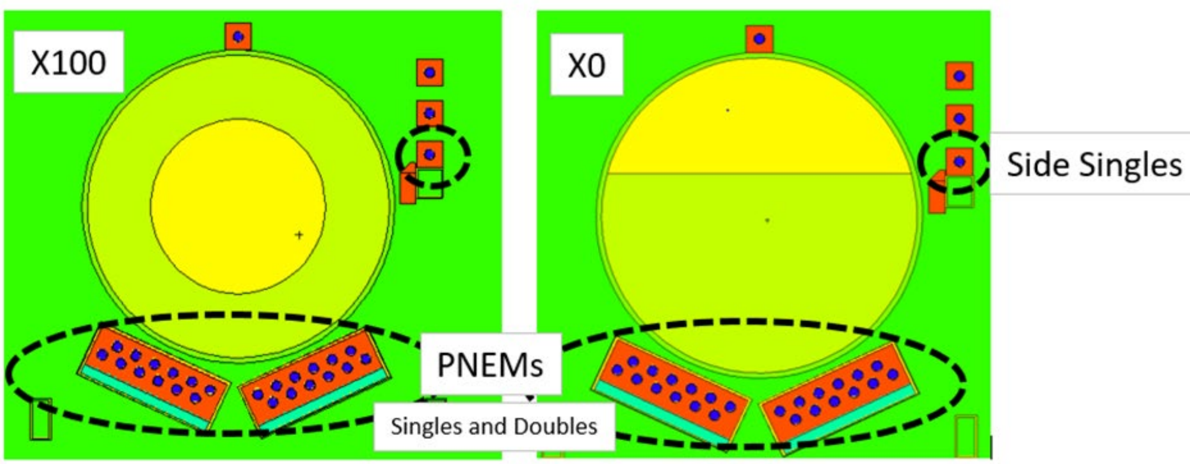

Figure 2 MCNP modelling of detectors (2 briefcase size polyethylene pods (12 x10 atm 3He detectors). Enrichments, ^{235}U-to-^{234}U ratio; UF_6 distribution (X-factor, percentage of UF_6 bound to walls).

Two additional studies are underway. One will determine the usefulness of the gamma/neutron ratio from (α,n) reactions for nondestructive analysis measurement systems. The second will examine the magnitude of the neutron source term from molten salts and if the uncertainties in the source term have an impact to reactor calculations and safeguards measurements.

Improvements to the (α,n) libraries will reduce uncertainties in NDA measurements and simulations for safeguards applications including UF_6 assays, SNM neutron measurements and monitoring of advanced fuels. The impact of nuclear data uncertainties on UF_6 cylinder assays and the usefulness of gamma/neutron ratios for nondestructive analysis of SNM paid particular attention to the $^{19}F(\alpha,n)^{22}Na$ reaction. Other applications of interest are the passive neutron and gamma source terms from special nuclear materials and advanced reactor fuels.

It is recommended that a new ENDF sublibrary be established that includes neutron spectrum, gamma emission and covariance data. To accomplish this, new experiments are required that provide information on the excited states of the compound nucleus in order to inform both the cross sections and the theoretical calculations of the neutron emission spectra, and new evaluations of the relevant isotopes will need to be performed. Thick target and total neutron yield measurements are required to normalize the data and ensure consistency between energy differential and integral data.

Priority isotopes include the following:

- For current safeguards: ^{19}F, $^{17,18}O$
- For nondestructive analysis of actinides compounds: $^{6,7}Li$, ^{9}Be, $^{10,11}B$ ^{13}C, $^{14,15}N$, $^{17,18}O$, ^{27}Al
- For advanced reactor fuels and safeguards: $^{6,7}Li$, ^{9}Be, $^{10,11}B$ ^{13}C, $^{14,15}N$, $^{17,18}O$, ^{19}F, ^{23}Na, $^{25,26}Mg$, $^{27,29,30}Si$, ^{37}Cl and ^{41}K.
- For low background measurements, cross sections are needed up to 10 MeV on all elements with the priorities determined by the specific application.
- Priority reactions for radionuclide production at energies up to 30 MeV: $^{69,71}Ga(a,n)$, $^{nat,40}Ca(a,n)$, $^{58}Ni(a,n)$, $^{nat}In(a,2n)$

ACTIVE INTERROGATION SCOPING STUDY

The active interrogation scoping study examined uncertainties in techniques that rely on gamma emissions resulting from neutron inelastic scattering and capture in objects of interest during interrogation with 14 MeV neutrons. These techniques have been explored for applications that require the detection of specific elements, such as threat and contraband detection, coal analysis, and oil exploration. Incident neutrons from a source interact with the contents of an unknown item or a material stream and can induce gammas specific to the isotopes present within the item or stream. For example, neutron inelastic scattering on ^{16}O can induce a 6.13 MeV gamma ray for detection, and neutron capture on ^{14}N can induce a 10.8 MeV gamma ray for detection. Along with the 4.43 MeV gamma ray from inelastic scattering on ^{12}C, these signatures can be considered for a bulk explosives detection application [1]. A similar isotopic analysis can be performed for chemical agents [15], drugs [16], coal [17], hydrocarbons in oil deposits [18], or other materials of interest, especially for the purpose of distinguishing them from benign materials that may also be part of a material stream. Using an appropriately intense neutron source and array of gamma detectors, the minimum capability enables material identification while knowing the emission time and direction of each source neutron enables an additional capability with a material distribution assessment.

The research and development lifecycle for applications that employ neutron-induced gamma spectrometry typically involves radiation transport modeling. The modeling may have at least one of the following objectives: (1) to explore relative intensities of gamma signatures, (2) to devise methods to extract the signal and estimate the background, (3) to predict the measurement time required to observe the signal, (4) to iterate the system design to yield the maximum signal-to-noise, or (5) to compare model predictions to measured data. The ability to achieve these objectives depends upon the quality of the nuclear data and the fidelity of the modeling codes.

This scoping study has identified shortfalls in the nuclear data and modeling tools that adversely affect the radiation transport modeling phase of the lifecycle. Examples of shortfalls have been identified in oil exploration where radioactive sources of neutrons or D-T neutron generators are used with NaI, BGO, GSO or LaBr detectors. Per Reference [18]], the discrepancies in modeling and experiment can be traced to cross-section tables within the Evaluated Nuclear Data File (ENDF) libraries, because the physics capability built into MCNP [19] has remained largely unchanged across recent ENDF releases. Some cross-section tables in the ENDF libraries have declined in quality (e.g., Fe, Ca) or are incomplete (e.g., Ba, La, Br, Ce). However, the full extent of the shortfalls is less well understood in research areas with observables involving energy and angular correlations. One ongoing research effort involves developing 3-D reconstruction algorithms for material identification using an associated particle imaging (API) D-T neutron generator and a pixelated array of plastic scintillator [20]. An example of a measurement and modeling discrepancy for three annuli composed of high-density polyethylene, steel, and tungsten is shown in Figure 3. The Geant4 model overestimates the inelastic scattering gamma counts by a factor of 2.6 for the HDPE annulus, 2.8 for the steel annulus, and 1.9 for the tungsten annulus. The time-of-flight observables in the measurement example are strongly dependent upon the accurately modeling the physics (energy, angle, time, particle number) of each inelastic reaction.

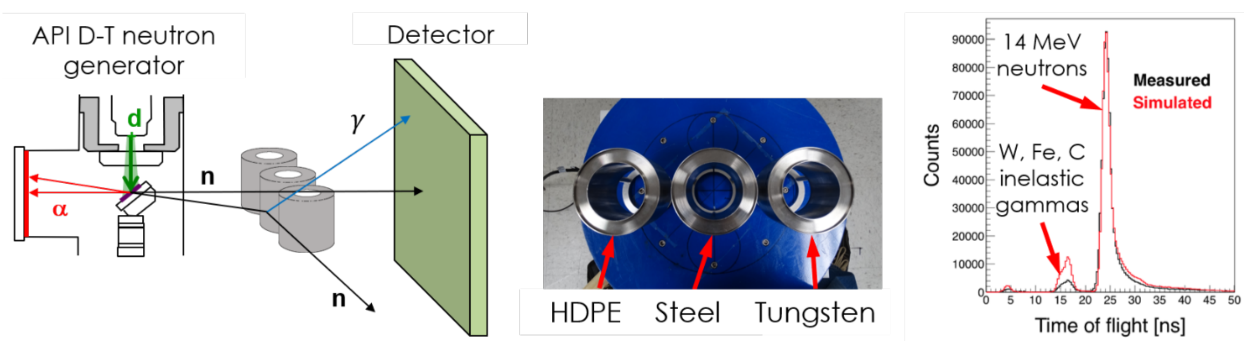

Figure 3 Example measurement and modeling discrepancy for three annuli composed of high-density polyethylene, steel, and tungsten. The Geant4 model overestimates the observed gammas for all three annuli: HDPE by a factor of 2.6, steel by a factor of 2.8, and tungsten by a factor of 1.9.

At present, the commonly used 3-D radiation transport codes of Geant4.10 and MCNP6.2 have known modeling shortfalls that prevent accurate physics modeling of inelastic scattering. MCNP6.2 does not emit neutrons in coincidence with de-excitation gammas without the Hauser-Feshbach solver CGM, which is too slow for practical use. Geant4/NeutronHP emits neutrons and gammas in coincidence when the data is formatted properly but ignores the breakup flags, leading to the emission of unphysical gammas. Improving the physics modeling within these codes will also require improvements to the nuclear data libraries. The ENDF/B nuclear data libraries, which currently store data in the ENDF-6 format, provide limited and sometimes inaccurate information on the gamma branching ratios needed to model a gamma cascade.

To address the modeling and nuclear data shortfalls, a research roadmap has been drafted with the following efforts:

a. Improve the nuclear data libraries by (1) resolving inconsistencies between ENDF and ENSDF, (2) beginning the transition from the ENDF-6 format to new General Nuclear Data Structure (GNDS) format, (3) making the Baghdad Atlas into a benchmark, and (4) incorporating new measurement data when available.
b. Improve the modeling capability by (1) porting the Monte Carlo General Interaction Data Interface (MCGIDI) [21] to C++, (2) extend the MCGIDI options to include a fast model-based fast event generator based upon RAINIER [22], and (3) integrate the result as a new Geant4 package.
c. Perform benchmark experiments, namely (1) integral experiments with DT API neutron generator, (2) differential experiments at GENESIS [23] and (3) neutron capture experiments using the MAD spectrometer at University of Massachusetts/Lowell.
d. Compare simulation results to experimental data to highlight discrepancies where new experimental data must be taken.

A list or priority elements has been developed for the research roadmap based upon the mission spaces that could benefit from modeling and nuclear data improvements. Priority elements were identified according to the categories of interest, namely

1. Structural (e.g., aluminum, steel, 3D printing materials),
2. Intervening/shielding/surrounding (e.g, polyethylene, water, thermal-neutron absorbers, lead, tungsten, concrete),
3. Detectors (e.g., organic scintillator, inorganic scintillator, semiconductor, detector housing, photomultipliers),
4. Source (e.g., housing including internal features and source reaction elements), and
5. Controlled substances (e.g., explosives, drugs, chemical agents, special nuclear materials).

The priority elements tentatively are H, C, N, O, Na, Al, Si, Fe, Cu, Pb, W, U, and Pu. The next tier priority elements are He, Li, Be, B, Si, Cl, Cr, Mn, Ni, Ge, Br, Cd, I, Cs, La. The elements completing the list are F, Mg, P, S, Ar, K, Ca, Ti, As, Kr, Mo, Sn, Sb, Xe, Gd, Bi, Np, Am.

CONCLUSIONS

Two research programs were developed that include four national laboratories and three universities and span five years. The primary goal of the (α,n) nuclear data program is the reduce the uncertainties in nondestructive analysis important to nonproliferation and to ensure that the codes have access to a new ENDF evaluated nuclear data sublibrary. The (α,n) source term is important for measurements where actinides are in contact with light materials including fluorides, oxides and molten salts. The current rendition of the widely used SOURCES4C code would benefit from modernization including a new ENDF sublibrary and a mechanism for validation.

The primary goal of the neutron-induced gamma production program is to establish a capability to accurately and quickly simulate correlated neutron scattering and gamma-ray production in support of neutron and gamma-ray transport modeling. This capability is expected to be beneficial across multiple mission spaces that may have a growing interest in material identification using neutron-induced gamma emissions, especially as on-going neutron generator and detector development efforts realize their objectives over the next several years.

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